Quartett formation at (100)/(110)-interfaces of d-wave superconductors

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Across a faceted (100)/(110) interface between two $d_{x^2-y^2}$-superconductors the structure of the superconducting order parameter leads to an alternating sign of the local Josephson coupling. Describing the Cooper pair motion along and across the interface by a one-dimensional boson lattice model, we show that a small attractive interaction between the bosons boosts boson binding at the interface – a phenomenon, which is intimately tied to the staggered sequence of 0- and π-junction contacts along the interface. We connect this finding to the recently observed $h/4e$ oscillations in (100)/(110) SQUIDS of cuprate superconductors.

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The $d_{x^2-y^2}$-symmetry of the superconducting state in high-T_c cuprates causes a wealth of new phenomena at surfaces, grain boundaries or interfaces in these materials. In particular, the sign change of the order parameter around the Fermi surface is the origin of the most compelling experimental evidence for the d-wave nature of superconductivity in cuprates, as became manifest in the observation of half-flux quanta at interfaces on tricrystal substrates [1,2]. Already prior to these experiments it was recognized that conventional Josephson junctions (0-junctions) as well as π-junctions with a sign reversal of the Josephson coupling [3] can be realized in contacts between cuprate superconductors depending on the mutual orientation of their crystal lattice and the attached four-fold symmetry of the order parameters.

In (100)/(110) interfaces or grain boundaries of d-wave, cuprate superconductors the CuO_2 lattices meet at 45 degrees, such that the $d_{x^2-y^2}$-order parameter lobes of the two superconductors point from a nodal towards an antinodal direction (see also Fig. 1). If the interface were perfectly flat, no net tunneling supercurrent would therefore flow. Microscopic roughness, however, allows for local supercurrents across interface facets [4]; the current direction at each facet is thereby determined by the relative phase of the clover leave lobes pointing towards the facet's surface. This special situation at (100)/(110) interfaces has led to a variety of effects like spontaneous supercurrent loops [4], locally time-reversal symmetry breaking phases [5,6], or anomalous field dependencies of the critical current density [7]. Yet another peculiar experimental observation was recently reported for SQUIDs with (100)/(110) interfaces, where the flux periodicity of the I-V characteristics was found to be h/4e, i.e. half a flux quantum [11]; this finding is the motivation for the present work, in which we propose a possible mechanism for pair binding or quartett formation in the interface.

Networks of Josephson junctions in array geometries or even granular superconductors are conveniently modelled by classical XY- or extended quantum phase Hamiltonians [8]. These models in fact can be derived from a purely bosonic description for the Cooper pair tunneling processes, if fluctuations in the bulk of the superconducting order parameter can be neglected [9]. By this means the boson kinetic energy translates directly into the Josephson coupling energy of the quantum phase Hamiltonian. The boson formulation furthermore allows for the advantage, that in the hard-core limit an exact mapping to a spin-1/2 Hamiltonian is possible [10], so that preexisting knowledge for the spin model can be transferred to the boson problem.

In this Letter we follow the latter strategy to analyze a bosonic lattice model Hamiltonian with a staggered sign for the hopping amplitude representing an alternating sequence of superconducting 0- or π-junctions. We show that the special staggered structure of the kinetic energy term strongly enhances the tendency towards boson pair formation in the presence of a weak attractive interaction, as revealed by the formation of bound triplets in the groundstate of the equivalent spin Hamiltonian. In a closed loop Aharonov-Bohm SQUID geometry of the underlying boson model, oscillations with a flux periodicity h/q are therefore expected, where q is the total charge of a boson pair, i.e. an electronic quartett. We interpret our results as a hint for a possible and intriguing alternative explanation of the observed $h/4e$ oscillations in high-T_c SQUIDS with (100)/(110) interfaces [11].

We start from the geometry shown in Fig. 1 and translate it into the Hamiltonian

$$H = \sum_{\alpha i} \left(-t(-1)^i a^+_{\alpha i+1} a_{\alpha i} - t' a^+_{\alpha i+2} a_{\alpha i} + h.c. \right) + H_\Phi + \sum_{\alpha i} \left[V a^+_{\alpha i} a_{\alpha i} a^+_{\alpha i+1} a_{\alpha i+1} + U \left(a^+_{\alpha i} a_{\alpha i} - 1 \right) a^+_{\alpha i} a_{\alpha i} \right] \quad (1)$$

with boson creation and annihilation operators $a^+_{\alpha i}$ and $a_{\alpha i}$ and $H_\Phi = -t_\perp \sum_j a^+_{1j} a_{2j} e^{i\Phi(-1)^j/2} + h.c.$. In the disk-shape geometry in Fig. 1 the (100)/(110) interface between the two d-wave superconductors is represented by

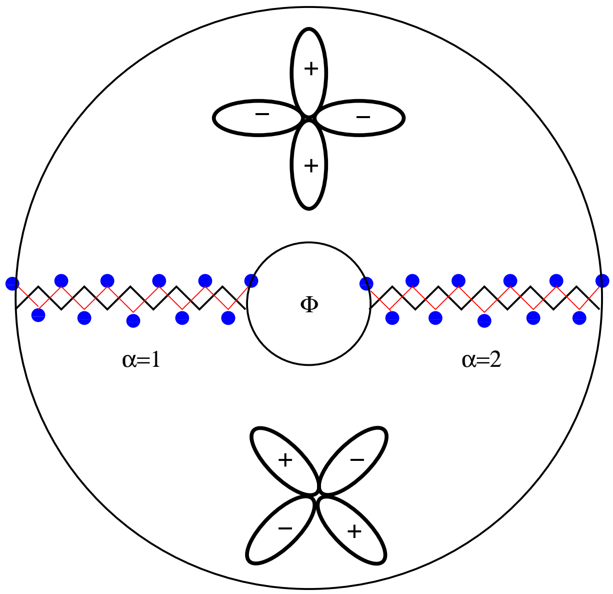

FIG. 1. SQUID geometry for two $d_{x^2-y^2}$-superconductors with two (100)/(110) interface contact regions (labelled $\alpha = 1, 2$) represented as bold saw-tooth lines. The thin zig-zag line, which crosses the interface sections and connects the circles, defines the chain for the model Hamiltonian Eq. (1).

a saw-tooth line - assuming that the interface splits into a regular sequence of orthogonal facets. In a dc-SQUID setup a magnetic flux Φ may pass through the hole in the disk center, which separates the two interfaces labelled by $\alpha = 1, 2$. The circles mark chain sites, between which bosons (Cooper pairs) can hop with or without crossing the interface. The latter next-nearest-neighbor processes have the unique sign $-t'$ for their hopping amplitude, while the former processes have an amplitude with an alternating sign due to the misalignment by $45°$ of the $d_{x^2-y^2}$-wave order parameter lobes on both sides of the interface. In Eq. (1) U and V denote the onsite and nearest-neighbor interaction strengths; in the following we will in particular explore the effect of a weak attraction $V < 0$. The two interfaces $\alpha = 1, 2$ are connected by $t_\perp$, which contains the phase factor of the threading flux Φ. If boson (Cooper pair) binding occurs in the interface, oscillations with flux periodicity $h/4e$ are expected.

A phase change for the boson operators at every second pair of adjacent sites according to $b^+_{\alpha 4i} = -a^+_{\alpha 4i}$, $b^+_{\alpha 4i+1} = -a^+_{\alpha 4i+1}$, $b^+_{\alpha 4i+2} = a^+_{\alpha 4i+2}$, $b^+_{\alpha 4i+3} = a^+_{\alpha 4i+3}$ transforms the kinetic energy part of the Hamiltonian for each interface into

$$H_{kin} = \sum_{\alpha i} \left[-t b^+_{\alpha i+1} b_{\alpha i} + t' b^+_{\alpha i+2} b_{\alpha i} + h.c. \right] ; \qquad (2)$$

all other terms remain unchanged. Importantly, for a sequence of ordinary 0-junctions the second term in Eq. (2) appears with a negative sign.

We now focus on the physics in *one* interface. As anticipated above we consider the hard-core limit $U \to \infty$, in which the boson problem maps onto a spin-1/2 model by means of the transformation [10]

$$S^+_i = (-1)^i b_i,\; S^-_i = (-1)^i b^+_i,\; S^z_i = \frac{1}{2} - b^+_i b_i \,. \qquad (3)$$

The resulting spin Hamiltonian reads

$$H_S = \sum_i \Big[\quad J_1 \left(S^x_i S^x_{i+1} + S^y_i S^y_{i+1} + \Delta S^z_i S^z_{i+1} \right) + J_2 \left(S^x_i S^x_{i+2} + S^y_i S^y_{i+2} \right) \Big] \qquad (4)$$

with the spin exchange coupling constants $J_1 = 2t$, $J_2 = 2t'$, and the anisotropy parameter $\Delta = V/2t < 0$. This model – including an additional anisotropy of the next-nearest-neighbor exchange J_2 – has been studied before in the context of metamagnetic transitions [12]. In particular, analytical results for pairing of magnons (bosons in the original language) were derived, and also a tendency to form clusters of more than two particles was obtained for certain parameter regimes. Because of its relevance for the quartett formation, we start to discuss the binding problem in the original bosonic language.

From the insight into the physics of the spin-chain model we infer, that for $t' > 0$ the partial frustration of the kinetic energy favors the binding of bosons, which represent the Cooper pairs. For each total momentum K of a pair of bosons, the bound state can be written as

$$|\psi_K\rangle = \sum_{j>0} A_j \sum_n e^{-\mathrm{i}K(n+j/2)}\, b^+_{n+j} b^+_n \,. \qquad (5)$$

The Schrödinger equation for the bound state, $H_1|\psi_K\rangle = \lambda_K|\psi_K\rangle$, with the Hamiltonian H_1 of one interface can be solved with the following ansatz

$$A_j = (\gamma_1)^j - (\gamma_2)^j , \qquad (6)$$

where γ_1 and γ_2 are the two solutions of the equation

$$-2t \cos\frac{K}{2} \left(\frac{1}{\gamma} + \gamma \right) + 2t' \cos K \left(\frac{1}{\gamma^2} + \gamma^2 \right) = \lambda_K \,, \qquad (7)$$

with $|\gamma_{1,2}| < 1$; the eigenvalue λ_K has to satisfy

$$\lambda_K = V + 2t' \cos K (\gamma_1^2 + \gamma_2^2 + \gamma_1\gamma_2 + 1) - 2t \cos\frac{K}{2} (\gamma_1 + \gamma_2) \,. \qquad (8)$$

The size of the pair is determined by the quantity $\xi = -1/\ln[\max(|\gamma_1|, |\gamma_2|)]$ and decreases with increasing V.

The critical interaction V_b for binding is determined by the condition, that the minimum of λ_K with respect to all possible pair momenta K equals twice the minimum of the one-particle energy $E_k = -2t \cos k + 2t' \cos(2k)$. The wave vector, which leads to the minimum E_k is $k_{\min} = 0$ for $\alpha = t'/t \leqslant 1/4$ and $k_{\min} = \arccos[1/(4\alpha)]$ for $\alpha \geqslant 1/4$. In our analysis we find that the optimum two-particle wave vector is $K_{\min} = 0$ for $\alpha \leqslant 1/(2\sqrt{2})$ in agreement with previous results for the spin-chain model [12], and

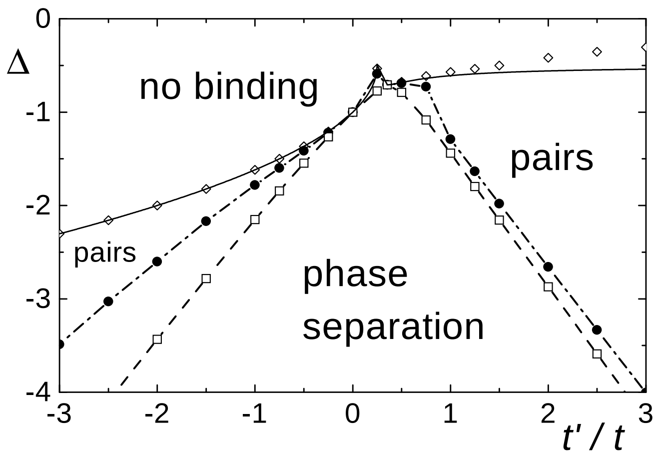

FIG. 2. Phase diagram of the interface model according to the number of particles n, which form bound composites; $\Delta = V/2t$. The full line corresponds to the analytical solution Eq. (9). Open diamonds indicate the pair binding boundary, full circles correspond to the transition from $n = 2$ to $n > 2$, and open squares denote the onset of phase separation.

$K_{\min} = 2k_{\min}$ for $\alpha \geqslant 1/(2\sqrt{2})$. The results for the minimum attraction necessary for binding can be summarized as follows with $\Delta_b = V_b/2t$:

$$\Delta_b = \begin{cases} -\dfrac{1+\sqrt{1-4\alpha}}{2} & \text{for } \alpha = t'/t \leqslant 1/4, \\ -2\alpha & \text{for } 1/4 \leqslant \alpha \leqslant 1/(2\sqrt{2}), \\ -\dfrac{1}{8\alpha}(\sqrt{16\alpha^2-1}+1) & \text{for } \alpha \geqslant 1/(2\sqrt{2})\,. \end{cases} \tag{9}$$

This function is represented by the full line in Fig. 2. Clearly, a small to moderate attraction is enough to lead to pair binding for positive t', which represents the alternating sequence of 0- and π-junctions, particularly for small hopping amplitudes t across the interface. Specifically, for the physically reasonable regime $t'/t > 1$ an attractive interaction of order t is sufficient for boson-pair formation; the energy scale for t should be determined by the Josephson coupling energy. Although ξ is very sensitive to V and diverges for $V \to V_b$, typical pair sizes for $V \sim t$ and $t' > 2t$ are an order of magnitude larger than the size of an individual facet.

It is known, particularly in models with strong correlations, that pairing competes with phase separation [13] and the tendency to bind in groups of more than two particles. To explore these possibilities we have studied numerically the equivalent spin Hamiltonian Eq. (4) in a chain of $L = 16$ sites. For each total spin projection S_z, which translates into a number of flipped spins (i.e. magnons) $m = L/2 - S_z$ added to the fully polarized ferromagnetic ground state, we have calculated the ground-state energy $E(m)$. To minimize finite-size effects and to accurately obtain the energy for one magnon, it is necessary to minimize over twisted boundary conditions [12]. If the particles in the system (bosons in the original language or magnons in the spin language) prefer to bind in groups of n particles, the quantity $e(m) = (E(m) - E(0))/m$ is minimized for $m = n$. We argue that phase separation occurs, when the condition $E(m) > (mE(L) + (L-m)E(0))/L$ holds for all m.

In Fig. 2 we show the resulting ground-state phase diagram. $\Delta = V/2t$ is the measure for the strength of the attractive interaction and $\alpha = t'/t$ is the ratio of hopping amplitudes for the motion along and across the interface. $\alpha > 0$ represents the alternating sequence of 0- and π-junctions, while π-junctions are absent for $\alpha < 0$. Four different regions are indicated in Fig. 2: the strong attraction regime, in which there is phase separation, a regime without binding and two intermediate phases, in which the size of the optimum particle cluster is $n = 2$, or $n > 2$. In the latter region n increases in unit steps as the attraction increases, except for $t' > t$, where only even n appear. The asymmetry between positive and negative t' with respect to the stability of boson-pair binding is evident, underlining the importance of the existence of π-junctions in the quartett formation. The numerical results for the border between $n = 1$ and $n = 2$ are in excellent agreement with the analytical results of Eqs. (9) – except for $t' > t$, where finite-size effects are present.

Comments remain in order about a possible origin of an attractive interaction for the bosons (i.e. Cooper pairs) in cuprate superconductors (or superconductors in general). We first note that the idea of quartett formation has been put forward before in nuclear physics [14]; proposals exist, that four-particle condensation may occur as a phenomenon alternative or complementary to nucleon pairing [14]. In cuprates, the possible existence of clusters of pairs has been discussed within the mesoscopic Jahn-Teller pairing model [15]. It was furthermore proposed, that due to strong phase fluctuations cuprates may be close to an exotic superconducting phase with quartet condensation [16].

A viable mechanism for electron pairing in high-T_c superconductors arises from antiferromagnetic (AF) spin fluctuations in a doped Mott-insulating host [17]. While a nearest-neighbor electron-electron attraction is dynamically generated from a local repulsive Coulomb interaction, a correlation of the pair motion in an environment with short range AF order is expected to optimally minimize the pair motion induced breaking of AF bonds. A simple picture for the source of binding in a system with short range AF correlations is obtained thinking in terms of static holes added to a Néel antiferromagnet on a square lattice: if two separated holes are added, they break 8 bonds. If instead they are added as nearest neighbors, only 7 bonds are broken. Naturally, this argument can be extended to more particles, suggesting that the binding mechanism may be active also for more than two particles and that the actual size of the composite object is determined by the competition with the kinetic energy of the particles, in a similar way as it happens in our bosonic model for a single interface.

Contrary to the few-particle problem in nuclear

physics, four-particle interaction vertices have so far been unexplored in correlated electron lattice models with superconducting instabilities, and therefore no firm basis exists for a discussion of correlated pair motion or even quartett formation tendencies. Yet, it is well established, that the quantitative description of the spin dynamics in undoped cuprates requires to include a sizable ring-exchange coupling [18], which naturally arises in strong-coupling expansions in next to leading order around the atomic limit [19]. A ring-exchange coupling does indeed contain 4-particle interactions between electrons on plaquettes of a square lattice. The complex structure of such an interaction has not been analyzed and its consequences for the dynamics of doped holes or the pair-wavefunction in the superconducting state remains unknown.

In the present analysis we have assumed a small attractive pair interaction. It is likely that such a weak interaction does not lead to observable phenomena in the bulk of a correlated superconductor; but at the peculiar (100)/(110) interface between d-wave superconductors seemingly subdominant 4-particle correlations may lead to new pairing tendencies and the possibility for quartett formation. In fact, one may argue, that the binding tendency is enhanced at the interface, because the cost in kinetic energy is reduced due to the phase factors of the d-wave pairs and the concomitant staggered sign of the hopping amplitudes in the bosonic language.

In the context of frustrated Josephson junction networks an alternative mechanism of Cooper-pair binding, based on a Z_2 symmetry of a particular geometry was reported for Aharonov-Bohm cages [22]. In this case 0- and π-junctions are realized on plaquettes, which are threaded by one flux quantum. In a one-dimensional arrangement these plaquettes are interconnected in a geometry, which leads to perfectly flat bands and thus to particle localization. Interactions may then lead to delocalized two-particle bound states or mobile charge $4e$ composite objects, which in closed loop SQUIDs should also give rise to an elementary $h/4e$ period of flux. A common feature of this proposal and the mechanism discussed in this Letter is indeed the important role of the partial frustration of kinetic energy.

If the mechanism discussed in this letter is indeed at work at (100)/(110) interfaces, the experimental observation of $h/4e$ flux periodicities in (100)/(110) SQUIDS of high-T_c superconductors would follow as a natural consequence. Yet, as discussed in [11], more conventional proposals of a suppressed $\sin\varphi$ component and a dominant $\sin 2\varphi$ component of the Josephson current at the (100)/(110) interfaces are available and present a vivid alternative to explain the experimental findings [20,21]. We believe, however, that the above discussed mechanism of quartett formation offers an intriguing new route for so far unexplored pair-binding phenomena in superconductors [23].

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